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Radiocarbon-based source apportionment of elemental carbon aerosols at two South Asian receptor observatories over a full annual cycle

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Abstract

Black carbon (BC) aerosols impact climate and air quality. Since BC from fossil versus biomass combustion have different optical properties and different abilities to penetrate the lungs, it is important to better understand their relative contributions in strongly affected regions such as South Asia. This study reports the first year-round ^{14}C-based source apportionment of elemental carbon (EC), the mass-based correspondent to BC, using as regional receptor sites the international Maldives Climate Observatory in Hanimaadhoo (MCOH) and the mountaintop observatory of the Indian Institute of Tropical Meteorology in Sinhagad, India (SINH). For the highly-polluted winter season (December–March), the fractional contribution to EC from biomass burning (f_{bio}) was 53 ± 5% ($n = 6$) at MCOH and 56 ± 3% at SINH ($n = 5$). The f_{bio} for the non-winter remainder was 53 ± 11% ($n = 6$) at MCOH and 48 ± 8% ($n = 7$) at SINH. This observation-based constraint on near-equal contributions from biomass burning and fossil fuel combustion at both sites compare with predictions from eight technology-based emission inventory (EI) models for India of $(f_{bio})_{EI}$ spanning 55–88%, suggesting that most current EI for Indian BC systematically under predict the relative contribution of fossil fuel combustion. A continued iterative testing of bottom-up EI with top-down observational source constraints has the potential to lead to reduced uncertainties regarding EC sources and emissions to the benefit of both models of climate and air quality as well as guide efficient policies to mitigate emissions.

1. Introduction

Black carbon (BC) aerosols have multiple negative impacts, including on human respiratory health (e.g., Janssen *et al* 2012) and on climate (e.g., IPCC 2013). The large uncertainties in BC emissions both with respect to absolute fluxes and the relative contribution from fossil versus biomass combustion sources complicate our ability both to accurately understand and model the multiple BC climate effects, as well as to efficiently mitigate emissions to reduce the health impact. Hence, a major challenge with respect to both climate and air quality aspects of BC and other aerosols are to assess and reduce the large uncertainties in existing BC emission inventories (EI) (e.g., Zhao *et al* 2011, Bond *et al* 2013). Comparisons between predictions based on bottom-up technology-based EI and estimates based on atmospheric observations suggest that EI-driven models underestimate the BC climate effects by a factor of 2–3 (Andreae and Ramanathan 2013, Bond *et al* 2013, Cohen and Wang 2014). Further, the relative contribution of

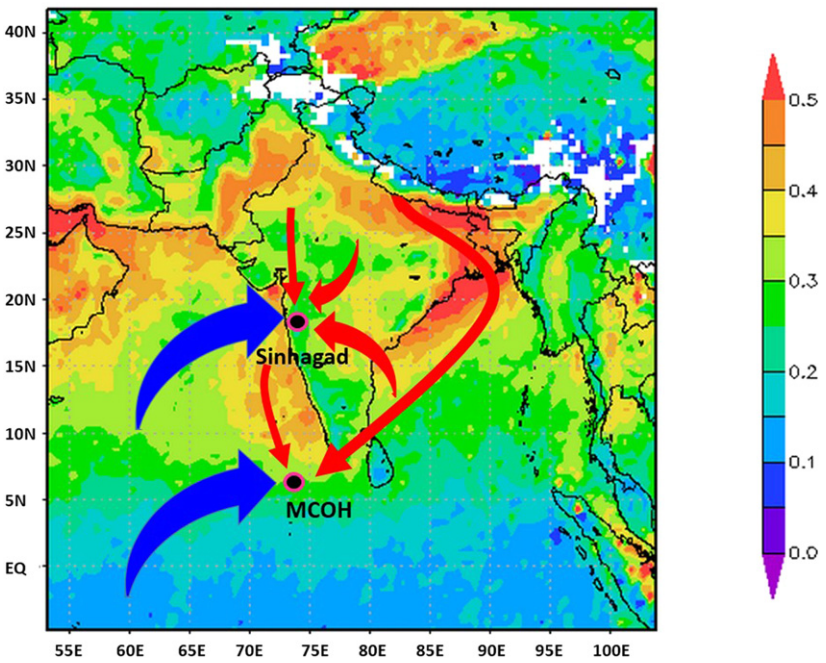

Figure 1. Map depicting average aerosol optical depth (AOD) at 550 nm during the January 2008 to April 2009 campaign. The locations of the Maldives Climate Observatory at Hanimaadhoo (MCOH) and Sinhagad Observatory (SINH) are shown. Coloured arrows indicate broadly the predominant BTs differentiated in two different clusters: dry season (red) and monsoon (blue). Individual and detailed back trajectory cluster analysis for this campaign is available in Sheesley *et al* (2012) and Kirillova *et al* (2013).

fossil versus biomass sources to BC, suggested by bottom-up EI, appear to systematically under-predict the fossil contribution relative to top-down source apportionment based on the source-diagnostic ^{14}C composition of BC aerosols in the actual atmosphere of both South and East Asia (e.g., Gustafsson *et al* 2009, Chen *et al* 2013). However, such observation-based source constraints have to this point been limited to shorter campaigns (weeks–month) and there is no single year-round ^{14}C-based assessment of BC sources for Asia or anywhere in the World.

Natural abundance radiocarbon ($\Delta^{14}C$) analysis is a powerful method for quantitatively differentiating between fossil versus biomass sources of carbonaceous aerosols in the actual atmosphere. Such information is important for diagnosing and reducing the current large uncertainties in EI of OC and BC aerosols. The present study provides, for the first time, measurements for a full-year cycle in Asia of ^{14}C of elemental carbon (EC)-the common thermal-optical mass-based correspondent to optical BC. Aerosol samples were collected at two well-established regional receptor sites in South Asia: Sinhagad (SINH) at a mountain-top in Western India and at the international Maldives Climate Observatory on the island of Hanimaadhoo (MCOH).

2. Methods and materials

2.1. Sampling locations and approach

Sampling was conducted at two regional receptor sites in South Asia (figure 1). The MCOH (latitude 6°78′N, longitude 73° 18′E, 15 masl) is located at the Northern tip of Hannimadhoo, a small island in the Republic of the Maldives in the Indian Ocean. The other site, Sinhagad (SINH, 18° 21′ N and 73° 45′ E, 1450 masl) is a rural-high altitude site on a mountaintop in the Western Ghat mountain ranges in Western India. Both MCOH (e.g., Corrigan *et al* 2006, Ramanathan *et al* 2007, Granat *et al* 2010, Sheesley *et al* 2012, Bosch *et al* 2014) and SINH (e.g., Momin *et al* 2005, Gustafsson *et al* 2009, Kirillova *et al* 2013, Budhavant *et al* 2014), are frequently used for studies of S. Asian aerosols. Samples were collected near-continuously at both sites during fifteen months in 2008–2009 (83% coverage at SINH and >99% coverage at MCOH), comprising two dry winter seasons, a monsoon season and the transition periods. The large mass of aerosol EC required to meet accelerator mass spectrometry detection limits for microscale ^{14}C measurements, were obtained using high-volume total suspended particle (TSP) samplers operated at 14–19 $m^3\,hr^{-1}$. The samples were collected on 140 mm quartz fibre filters (Tissuquartz filters from Pall Gelman) in custom-built filter holders as described earlier (e.g., Gustafsson *et al* 2009, Sheesley *et al* 2012, Kirillova *et al* 2013). The TSP approach (which collects all aerosols smaller than approximately 30 μm in aerodynamic radius) was used in this study as it was desired to assess the sources of the full population of aerosol EC, as opposed to using a finer cut-off such as $PM_{2.5}$ selecting mainly for respiratory particles. The sampling interval at MCOH was ~one week during the non-monsoon periods and ~two weeks during

monsoon seasons. The SINH site was maintained at a near-constant one-week sample duration. Filter blanks were collected approximately once per month for each site. Quartz fibre filters were pre-baked at 450 °C for 12 h and individually stored in aluminium foil envelopes in double Ziploc bags in the freezer. More details about sampling sites and operation as well as detailed air mass back trajectories for this campaign are described in earlier studies reporting on other aerosol carbon fractions (Sheesley *et al* 2012, Kirillova *et al* 2013).

2.2. Carbon aerosol mass concentration and isotope analyses

Organic carbon (OC) and EC concentrations and radiocarbon composition were measured using previously established methods. Quantification of OC and EC used a thermal-optical transmission analyzer (Sunset Laboratory, Tigard, OR, USA) using the National Institute for Occupational Safety and Health 5040 method (Birch and Cary 1996). The average of the concentrations of the field blanks was taken into account for the calculation of atmospheric concentrations (SINH, OC = 0.063 μg m^{-3}; MCOH, OC = 0.075 μg m^{-3} and EC = 0.001 μg m^{-3}). Triplicate analyses of laboratory standards and field reference material showed that the analytical uncertainties were less than 5%.

The isotope composition of other carbon aerosol fractions have earlier been measured and reported for this campaign, including for total OC and soot carbon (Sheesley *et al* 2012) as well as for water-soluble organic carbon (WSOC; Kirillova *et al* 2013). The current study reports on the important EC fraction, which was isolated for offline ^{14}C analysis by separation and cryogenic isolation of the CO_2 evolved from the EC peak, as described in detail by Chen *et al* (2013). The subsequent ^{14}C analysis was then conducted collaboratively with the US National Ocean Sciences Accelerator Mass Spectrometry Facility in Woods Hole, MA, USA as described earlier (e.g., Zencak *et al* 2007a, 2007b, Gustafsson *et al* 2009, Chen *et al* 2013). The radiocarbon data are reported as fraction modern (f_m) which is converted to the Δ^{14}C scale (Zencak *et al* 2007b). By constraining S Asia specific source end-member values for both biomass combustion of $\Delta^{14}C_{biomass} = +199‰$ (Gustafsson *et al* 2009) and a fossil fuel combustion end member of $\Delta^{14}C_{fossil} = -1000‰$, the fraction biomass ($f_{bio}$) of the EC may be established directly from the Δ^{14}C signature of the sample using the following mass-balance relation:

$$\Delta^{14}C_{sample} = f_{bio}\Delta^{14}C_{biomass} + \left(1 - f_{bio}\right)\Delta^{14}C_{fossil}. \quad (1)$$

The fraction EC contributed by fossil fuel combustion (f_{fossil}) is thus $1 - f_{bio}$.

A recent community inter-comparison of ^{14}C measurements on aerosol samples demonstrated comparable results for aerosol total carbon (Szidat *et al* 2013). There is a need for future such comparisons of different techniques and operational definitions for isolating the EC fraction for ^{14}C measurements. While there certainly are some uncertainties, the current top-down ^{14}C-based EC source apportionment study is based on the same commonly employed thermal-optical transmission method that is also used to determine EC emission factors for different combustion systems used in the EC EI (e.g., Bond *et al* 2004, 2013, Wang *et al* 2014). Hence, since the same operational definition of EC is used in both the bottom-up EI and the present top-down source apportionment this allows for a direct comparison between the two. To test the sensitivity of the obtained ^{14}C-based source apportionment results to a putative exchange in the isolation between method-induced pyrolysis of WSOC and ambient EC, a sensitivity analysis, based on instrument-generated pyrogenic C (PyrC) and our earlier reported ^{14}C-WSOC has been performed (supplementary information text S1 and table S2). The results suggest at most a moderate influence of such a hypothetical instrument-methodological process, well within the existing variability of the ^{14}C–EC data.

3. Result and discussion

3.1. Seasonal variations of EC and OC

The South Asian climate is governed by the monsoon system, with rainy summers and dry winter periods, and two transitional periods, the pre-and post-monsoon phases. The monsoon period is characterized by Southerly winds and, over land, by an elevated atmospheric boundary layer. In contrast, the dry period have on average Northerly winds and a shallower boundary layer. The onset of the different seasons depends on the passing of the Intertropical Convergence Zone, and is thus different for the two presently investigated sites: Sinhagad (SINH) and the MCOH.

As a result, the surface concentrations of carbonaceous aerosols show significant seasonal variations (Sheesley *et al* 2012, Kirillova *et al* 2013, Krishna *et al* 2013, Kompalli *et al* 2014) with elevated concentrations during the dry winter season (December–March) and lowest concentration during the summer monsoon season (June–September) with intermittent levels during the transition periods. Our 16 month continuous measurements are consistent with this pattern of seasonal variability in OC and EC concentrations at both MCOH and SINH (figures 2(A) and (C)). Higher mean concentrations of OC and EC were obtained in the dry season and lower mean concentrations were observed in the wet season. The EC concentrations of week-long samples varied in wide ranges of 0.03–0.54 μg m^{-3} for wet season and

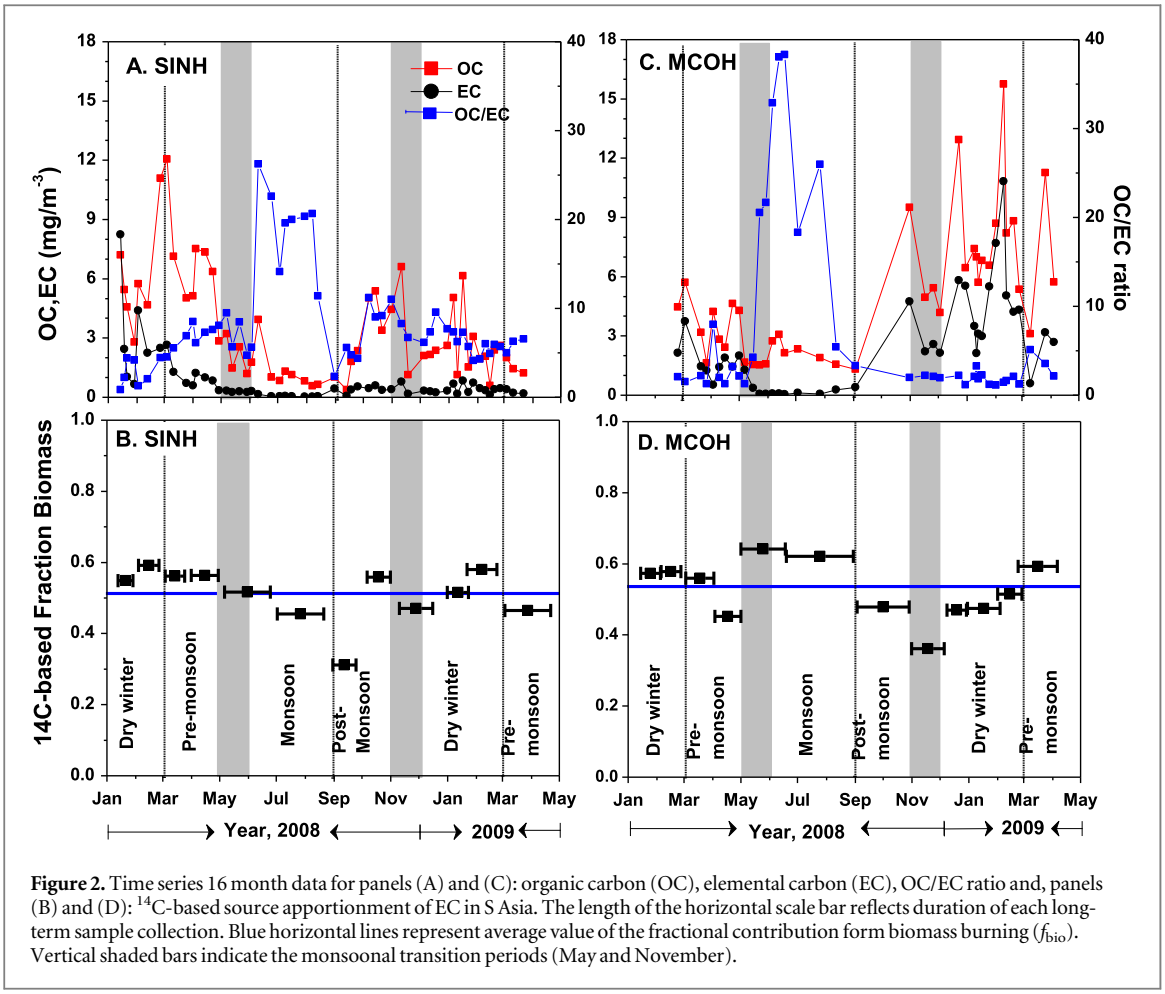

Figure 2. Time series 16 month data for panels (A) and (C): organic carbon (OC), elemental carbon (EC), OC/EC ratio and, panels (B) and (D): ^{14}C-based source apportionment of EC in S Asia. The length of the horizontal scale bar reflects duration of each long-term sample collection. Blue horizontal lines represent average value of the fractional contribution form biomass burning (f_{bio}). Vertical shaded bars indicate the monsoonal transition periods (May and November).

0.12–8.24 μg m^{-3} for dry season at SINH, and with 0.06–0.40 μg m^{-3} for wet season and 0.07–10.8 μg m^{-3} for dry season in MCOH.

A correlation between OC and EC concentrations may indicate similar source and geographic origin of carbonaceous particles. Significant correlations were observed between OC and EC both at MCOH (Pearson's correlation coefficient, $r^2 = 0.79$, $p < 0.0001$ for 39 samples) and at SINH ($r^2 = 0.34$, $p < 0.0001$ for 55 samples). These co-varying patterns suggested that the ambient concentration levels of carbonaceous species were controlled largely by processes such as primary source emissions and atmospheric dispersion rather than by secondary OC formation. This is consistent with findings by Kirillova *et al* (2013) for OC, and especially for WSOC, based on dual δ^{13}C–Δ^{14}C data for this same campaign. When split seasonally, OC and EC presented somewhat different correlations. At MCOH, they were highly correlated in all the seasons with r^2 value ranging from 0.57 ($p < 0.005$) to 0.59 ($p < 0.0001$), while at SINH they were strongly correlated in the dry winter season of 2009 with $r^2 = 0.83$ ($p < 0.0001$), but not distinctly correlated in the monsoon and dry season of 2008 with r^2 value ranging from only 0.18 to 0.20. The lower correlation coefficient during the SINH summer suggests contributions from different sources for OC and EC (e.g., biogenic secondary organic aerosol (SOA) to OC), effects of atmospheric processing on (primarily) OC or differential contributions from long-range versus regional emissions during the wet season.

3.2. OC/EC ratios

The mass ratio of OC to EC (OC/EC) reflects multiple processes in the atmosphere. (1) The OC/EC ratio is typically higher from biomass combustion than from fossil sources (e.g., Ram and Sarin 2010), (2) the OC/EC ratio is elevated by (mainly) biogenic SOA contributions (e.g., Saarikoski *et al* 2008), (3) the OC/EC ratio is affected by atmospheric processing (e.g., aging) of organic chemicals (e.g., Kroll *et al* 2011) and (4) the atmospheric lifetime for OC is shorter due to higher chemical reactivity and greater tendency for washout during rain events. Here, the OC/EC ratios ranged from 0.9 to 26 with an average of 8.0 ± 5.4 at SINH, and from 1.1 to 42 with an average of 7.4 ± 10 at MCOH (figure 2). The highest values were observed during the monsoon season for both sites (26.3 (13.7) for SINH and 38.3 (23.2) for MCOH, bracket values represents mean), whereas the lowest values were found during the dry winter period (0.88 (6.29) for SINH and 1.13 (3.51) for MCOH). These distinct seasonal trends suggests comparably larger contributions from biogenic SOA or pollen to OC during the

Table 1. Annual and seasonal source apportionment based on 16 month continuous ^{14}C–EC observations for Hanimaadhoo, Maldives (MCOH) and Sinhagad, India (SINH). (±) indicate the variability in the sampling.

	Number of samples	Fraction modern (F_m)	Standard deviation	Fraction biomass (f_{bio})
Sinhagad (SINH)				
2008 annual	9	0.61	0.09	51 ± 8%
Winter (December–March)[a]	5	0.68	0.03	56 ± 3%
Non-winter (March–November)[a]	7	0.58	0.10	48 ± 8%
March–April 2006[b]	3			54 ± 2%
Maldives Climate Observatory at Hanimaadhoo (MCOH)				
2008 annual	9	0.64	0.09	53 ± 9%
Winter (December–March)[a]	6	0.64	0.05	53 ± 5%
Non-winter (March–November)[a]	6	0.63	0.12	53 ± 11%
January–March 2006[b]	5			41 ± 4%
February–March 2012[c]	8			59 ± 4%

[a] Includes 2008 and 2009.
[b] Gustafsson *et al* (2009).
[c] Bosch *et al* (2014).

wet monsoon period, which is also expected from the warmer and wetter weather conditions during this phase, which favours biological activity (e.g., Genberg *et al* 2011).

3.3. Year-round source apportionment of EC aerosols

The $^{14}C/^{12}C$ characteristic of carbonaceous aerosol samples is a direct indication of the relative contribution of biomass (f_{bio}) and fossil fuel (f_{fossil}) combustion sources (equation (1)). The studied receptor sites were each influenced by seasonally-varying air masses with different geographical origins. Nevertheless, the f_{bio} varied over the 16 months of observations over a very similar range for both SINH (36–64%) and MCOH (31–59%) (figures 2(B) and (D) and SI table 1). At SINH, the total dry season (winter) average f_{bio} for EC was 56 ± 3% ($n = 5$; table 1). The remainder of the year (summer monsoon and transition periods) exhibited a mean f_{bio} for SINH EC of 48 ± 8% ($n = 7$; table 1). The winter season aerosol samples at SINH experienced influence by air masses from East/North East and Central India (detailed back trajectory cluster analyses shown in Sheesley *et al* 2012, Kirillova *et al* 2013). These times, the EC may be influenced from high emissions of aerosols due to biomass burning as suggested by the MODIS active fire counts' data (SI figure 1) that show higher incidences of fires over India and Bangladesh during the 2009 dry months. This may be due to burning of agricultural crop residues from primarily wheat and rice. Large-scale emissions from paddy-residue burning does however fall slightly outside the winter period during October–November and the same for wheat-residue burning in April–May. However, the timing of these activities are not absolute and they are ubiquitous features in the Indo-Gangetic Plain (IGP) (Badarinath *et al* 2009, Rajput *et al* 2014). The estimated emission budgets from the agricultural-waste burning on an IGP scale contributes a predicted ~22% of primary OC [252 ± 34 Gg y^{-1}] and 21% of EC [59 ± 2 Gg y^{-1}] (Rajput *et al* 2014). On the other hand, aerosols in the monsoon were associated with air masses primarily from Southwest i.e. from the Northern Indian Ocean and the Arabian Sea. The overall 2008-annual average value of f_{bio} at SINH was 51 ± 8% (table 1).

A similar source apportionment of EC between biomass versus fossil fuel combustion was observed at MCOH. There f_{bio} at MCOH was indistinguishable between the winter (53 ± 5%; $n = 6$; table 1) and non-winter seasons (53 ± 11%; $n = 6$; table 1). High f_{bio} at MCOH may be related to the fact that during the Southwest monsoon season; winds have been touching the Southeast coast of the African continent before passing through Indian Ocean and reaching the sampling location at MCOH. On a global scale, the largest open-burning emissions happen in Africa (Bond *et al* 2013). The major types of biomass burning in Africa include forest and savanna fires (Mkoma *et al* 2013). Field burning of agriculture residues and forest/wild fires occur in dry season (July–October) in Southern Africa and Madagascar (Schultz *et al* 2008, Mkoma *et al* 2013). Active fire spots (SI figure 1) were observed from MODIS satellite images during the dry season (wet season for MCOH) in June–October in Southern Africa and Madagascar, from where the air masses travelled more frequently to the sampling site at MCOH than to SINH. Hence, it is likely that biomass burning in SE Africa and Madagascar could have contributed to ambient carbonaceous aerosols at MCOH.

During dry season, air masses originated mainly in and around the Indian sub continent (figure 1), contributing to the high f_{bio} (EC) values at both stations.

This is consistent with the earlier picture of about equal contributions from fossil and biomass combustion during the high-aerosol impact from the highly polluted IGP spreading over great South Asian scales during dry periods (e.g. Lawrence and Lelieveld 2010, Sheesley *et al* 2012, Kirillova *et al* 2014). These year-round ^{14}C-based averages in EC source apportionment, suggest similar source attributions but with a somewhat smaller divergence in the biomass contribution to EC at the two regional sites than was apparent in the shorter 2006 campaign (Gustafsson *et al* 2009). These new winter results are inseparable from those reported for MCOH for a three-week high-intensity campaign in 2012 (Bosch *et al* 2014) (table 1).

3.4. Understanding the emission sources of carbonaceous aerosols in South Asia

Bottom-up EI of BC, which feed into climate and other atmospheric chemical-transport and health/air quality models, are challenged by large uncertainties related to both activity (tons fuel burnt) and especially the emission factor (kgBC/ton fuel burnt) for different sources (Zhao *et al* 2011, Bond *et al* 2013). These uncertainties, relating to the total amount of BC, also propagate into the estimates of the fractional contributions from different sources, e.g., fraction biomass (Chen *et al* 2013). The recent global high-resolution bottom-up EI for BC by Wang *et al* (2014) gives a $f_{bio} = 0.47$ for the Indian peninsula (defined as 6–32°N and 68–90°E) for the year 2007. This is significantly lower than the results from Bond *et al* (2004) of $f_{bio} = 0.75$ (for year 2000), the GAINS-IASA model ($f_{bio} = 0.70$, for 2010) and the results from Venkataraman *et al* (2005) for this region. However, the higher fossil contributions found by Wang *et al* (2014) are in better agreement with the year-round observationally-based ^{14}C-constrained source apportionment of the present study, with $f_{bio} = 0.53 \pm 0.08\%$ at MCOH and $f_{bio} = 0.51 \pm 0.08\%$ at SINH, which in turn also are consistent with previous shorter-term ^{14}C-source forensics estimates (Gustafsson *et al* 2009). It should be emphasized, however, that a direct comparison between bottom-up EI data and top-down constraints is also associated with some uncertainties owing to, for instance, the variability of air mass transport.

Taken together, the present and first-ever year-round study of ^{14}C–EC based source apportionment shows that both fossil and biomass combustion processes are about equally responsible for the emission of EC to the extensive Atmospheric Brown Cloud phenomena over South Asia. This highlights the importance of long-term campaigns, which gives a more comprehensive picture of the sources of climate-affecting aerosol carbon in this dynamically changing region.

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